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Influence of Substrate Temperature on Electrical Properties of PbO Thin Films Deposited by Chemical Spray Pyrolysis Technique

Abdulazeez O. Mousa, Ali F. Marmoss

Abstract :Nanostructured of (PbO) thin films were prepared by spray pyrolysis (CSP) technique at a different substrate temperatures (200, 250, 300, 350, 400, 450) ˚C . The films deposited were 177 nm thickness. It has been making electrical measurements such as D.C conductivity and, Hall effect for all films. The results showed that the conductivity of (PbO) increasing from [75.37 x 10^{-4} $(\Omega.cm)^{-1}$] to [339.99x10^{-4} $(\Omega.cm)^{-1}$] with increasing of substrate temperatures, as well as the results showed throughout the study that all films have tow activation energy and this energy increase with increasing of substrate temperatures. We noticed from the Hall effect measurements that the films have a negative Hall coefficient. This mean that the type of conducting (n–type charge carriers).

Keywords: Spray pyrolysis, lead oxide, electrical properties, activation energy, D. C. conductivity, Hall effect, thin film.

I. Introduction

Over the last few decades, research interest in the study of nanomaterials and their applications has been increasing, since these materials often demonstrate very different properties at the nanoscale level as compared to those at the macro level, such as new optical, magnetic, and electronic characteristics [1]. Furthermore, nanomaterials with high aspect-ratio structures and large surface areas offer exciting research possibilities because of such novel physical or chemical properties. As a result, the synthesis and characterization of one and two-dimensional metal oxide nanostructures have attracted considerable attention among the researchers. Among different chemical methods, spray pyrolysis technical is most popular today because its application to produce a variety of conducting and semiconducting materials [2]. The fundamental principle involved in the spray pyrolysis technical is the thermal decomposition of the compound to be deposited salts. Spray pyrolysis technique has a list of advantages. Spray pyrolysis technical is a simple and low cost to set up semiconductor thin films. Has the capacity to produce wide area, high quality films attached to standardized thickness [3].Transparent conducting oxide (TCO) thin films such as PbO, ZnO, SnO2, In2O3 and MoO3 have been studied in detail by many researchers [4]. These TCOs find extensive applications in thin film transistors, solar cells, phototransistors, optical storage devices, gas sensors, photo-thermal and photovoltaic conversions [5,6]. The difficulty of preparing exclusively single phase α-or β-PbO was pointed out earlier. The α-PbO was obtained earlier by pulsed laser ablation and spray pyrolysis [7]. The α-PbO was transformed to the meta-stable β-PbO when heat treated beyond 489 °C [8]. In the present work, we bring out a detailed investigation electrical properties on this film with $200 \leq T \leq 450$℃ prepared from spray pyrolytic decomposition of aqueous solutions of lead chloride .

II. Experiment

Lead Oxide thin films have been prepared by chemical spray pyrolysis (CSP) technique onto highly cleaned glass substrate with the dimensions (35× 25 ×1.35) mm^3. A homogeneous solution of (0.03M) was prepared by dissolving lead chloride compound ($PbCl_2.2H_2O$) by re-distilled water and a few drops of glacial acetic acid were then added to stabilize the solution. The solution was stirred for (1hr) with a magnetic stirrer, the temperatures used in this work were (200, 250, 300, 350, 400, 450) ˚C . The carrier gas was (compressed nitrogen) and the solution is fed into a sprayer nozzle at a pre-adjusted constant atomization pressure (4.5 bar) and we use (15 No. Of spray) as aconstant thichness (177) nm for all samples . The electrical conductivity has been measured as a function of temperature for films in the range (40 – 65) °C by using the electrical circuit. The measurements have been done using sensitive digital electrometer type Keithley (616) and electrical oven. The Hall effect was carried out according to the electrical circuit which contains a D.C. power supply with (0 – 40) volt and two digital electrometers (HMS-3000) to measure the current and

voltage. The resistivity (ρ) of the films is calculated by using the following equation [9] :

$$\rho = R \times A / L \quad (1)$$

Where R is the sample resistance, A is the cross section area of the films and L is the distance between the electrodes. The conductivity of the films was determined from the relation:

$$\sigma_{D.C} = 1/\rho \quad (2)$$

The activation energies could be calculated from the plot of $\ln\sigma$ versus 1000/T according to equation (3).

$$\ln \sigma_{D.C} = \ln\sigma_{\circ} - \Delta E / kT \quad (3)$$

Since : $\ln\sigma_{\circ} - \Delta E_{a} / k$ is numerical constant

III. 3. Results and Discussions

The electrical properties of prepared (PbO) thin films were investigated as follows:

D.C Electrical Conductivity

Fig. (1) show the variation of D.C conductivity (σ) vs. temperature (T), this figure show that (σ) increased with T, this is seems to be a normal behavior as one of semiconductor properties, due to the increasing of annealing temperature with increasing of substrate temperature.

The activation energy (E_a) obtained for these films is given in the Table (1), which calculated from the slope of $\ln\sigma$ vs. 1000/T, which shown in Fig.(2). From this figure the films having two activation energy depend on T value. This means there are two mechanisms for conductivity.The activation energy in the low temperature depends on the ionization impurity and at high temperature depends on the generation of electron-hole pairs. Table(1) shows that the value of Ea_2 is smaller than values of Ea_1 . This indicates that the conductivity depends on the temperature where $\sigma \propto T^{3/2}$ [10].

Hall Effect Measurements

The type of charge carriers, concentration (n_H) and Hall mobility (μ_H), has been estimated from Hall measurements. Table (2) shows the main parameters estimated from Hall effect measurements for PbO thin films deposited with different substrate temperatures in the range (200 – 450)˚C. We can notice from this Table that the films have a negative Hall coefficient. This mean that the type of conducting (n–type charge carriers), This results is in agreement with Zayed et al.[11].

Also we can notice from figures (3) and (4) respectively, that the carrier's concentration (n_H) increases with the increasing of substrate temperatures, while Hall mobility (μ_H) decreases with the increasing of substrate temperatures. Increasing the density of charge carrier's is essentially because of the lowering the potential barrier. While the decreasing of mobility is coming from the inverse relation between (μ_H) and (n_H). These results are agreement with Kang et al.[12].

Also From Table (2) the value of (σ) was increased with increasing substrate temperatures. That the structure of thin film was changed to decrease the grain size and decrease barrier potential of the internal grains, which make capture to the change carriers in the grain bounding and then increase the scattering. These results are in agreement which funded by [13].

It is observed from the Fig. (5) that the resistivity of PbO films decreases with increase in temperature, indicating the semiconducting nature of the as deposited samples. The resistivity decreases with increase in substrate temperature up to 350 °C, and then it starts increasing above 325 °C. A minimum resistivity value obtained for the film coated at 350 °C, which might be caused by the following two reasons: (i) least thickness obtained as a result of the removal of H_2O vapor, which might resist conduction between PbO grains. (ii) formation of vacancies of oxygen in the structure of the material, which serve as donors and contribute to higher conductivity. This is in accordance with Salunkhe et al. [14].

Conclusions

PbO thin films have been deposited on glass substrates at different substrate temperatures by the spray pyrolysis technique using lead chloride as the precursor salt. The role of substrate temperature on electrical

properties of the deposited films has been systematically investigated. (PbO) thin films were deposited on glass substrates as a function of different Substrate temperatures (T) ($200 \leq T \leq 450$)℃. All thin films had ohmic behavior. (PbO) thin films had n-type, low conductivity. Also, two activation energy. Mobility decrease with increasing of substrate temperatures. $\sigma_{D.C}$ was increased with increasing of Substrate temperatures.

References

[1] A. F Arulraj, D. Kanagarajan , P. Natarajan, J. Natarajan, "Study of oxidation behavior of lead thin films by thermal evaporation method " , International Journal of Current Research, (2014), vol.6, pp. 10140-10146 .

[2] P.Veluchamy, M. Sharon, M. Shimizu and H. Minoura. "Composition and photoactivity of lead oxide film prepared on a Pb electrode in the Pb/PbO potential region in 0.1 M NaOH + 0.1 M Na2SO4 at 80°C " , J. Electroanal. Chem, (1994), vol. 365, pp. 179-183.

[3] S.Ghasemi, M.F. Mousavi, M.Shamsipur and H. Karami.,"Sonochemical-assisted synthesis of nanostructured lead dioxide", Ultrason.Sonochem, (2008), Vol. 15, pp. 448-455.

[4]. L.Zhang, F. Guo, X. Liu, J. Cui and Y.T. Qian, "Metastable PbO crystal grown through alcohol-thermal process", J. Cryst. Growth., (2005), vol. 280, pp. 575-580.

[5] T. B.Light, J. M. Eldridge, J. W. Mattews and J. H. Greiner., "Structure of thin lead oxide layers as determined by x-ray diffraction", J. Appl. Phys., (1975), vol.46, pp. 1489-1492.

[6] Y.Pauleau and E. Harry, "Reactive Sputter-Deposition and Characterization of Lead-Oxide Films ", J. Vac. Sci. Technol. A, (1996), vol.14,pp. 2207-2214.

[7] M.Baleva, and V. Tuncheva, "Laser-assisted deposition of PbO films ", J. Mater. Sci. Lett, (1994),vol. 13, pp. 3-5.

[8] L.Madsen , D.and L. Weaver, "Characterization of lead oxide thin films produced by chemical vapor deposition", J. Am. Ceram. Soc, (1998),vol. 81, pp. 988-996.

[9] N. F. Mott and E. A. Davis, "Electronic Process in Non-Crystalline Materials", Clarendon Press, Oxford, (1979).

[10] H. A. Zayed, A M Abo-Elsoud, A. M. lbrahim and M. A. Kenawy, "Transport properties of $Sb_x\,Se_{1-x}$thin films", Journal of Physics D: Applied Physics, (1995), vol. 28, pp. 770-773.

[11] M. J. Kang , T. J. Park , D. Wamwangi, K. Wang , C. Steimer , S. Y. Choi and M. Wuttig," Electrical properties and crystallization behavior of $Sb_x\,Se_{100-x}$ thin films", Microsyst Technol, (2007), vol. 13, pp. 153-159.

[12] R. Kumaravel, V. Krishnakumar, K. Ramamurthi, E. Elangovan and M. Thirumavalavan, " Deposition of $(CdO)_{1-x}(PbO)_x$ Thin Films by Spray Pyrolysis Technique and Their Characterization", Journal of Thin Solid Films , (2007), vol.515, pp.4061– 4065

[13]. M. Suganya, A.R. Balu and K. Usharani, "Role of substrate temperature on the growth mechanism and physical properties of spray deposited lead oxide thin films", Mater. Sci. Poland, (2014), vol.32, p. 448.

[14] R.R. Salunkhe, D.S. Dhawale, T.P. Gujar, C.D. Lokhande," Structural, electrical and optical studies of SILAR deposited cadmium oxide thin films: Annealing effect ", Mater. Res. Bull., (2009),vol. 44, p. 364.

Table (1): D.C. conductivity parameters for (PbO) thin films at differentsubstrate temperatures.

Substrate temperatures (℃)	Ea_1 (eV)	Temp.range (K)	Ea_2 (eV)	Temp.range (K)	$\sigma_{R.T}\times10^{-4}$ $(\Omega.cm)^{-1}$
200	0.711	(313-323)	0.220	(328-338)	87.93
250	0.499	(313-323)	0.277	(328-338)	75.37
300	0.228	(313-323)	0.598	(328-338)	91.66
350	1.051	(313-323)	0.333	(328-338)	102.50
400	0.240	(313-323)	0.536	(328-338)	107.49
450	0.159	(313-323)	0.438	(328-338)	339.99

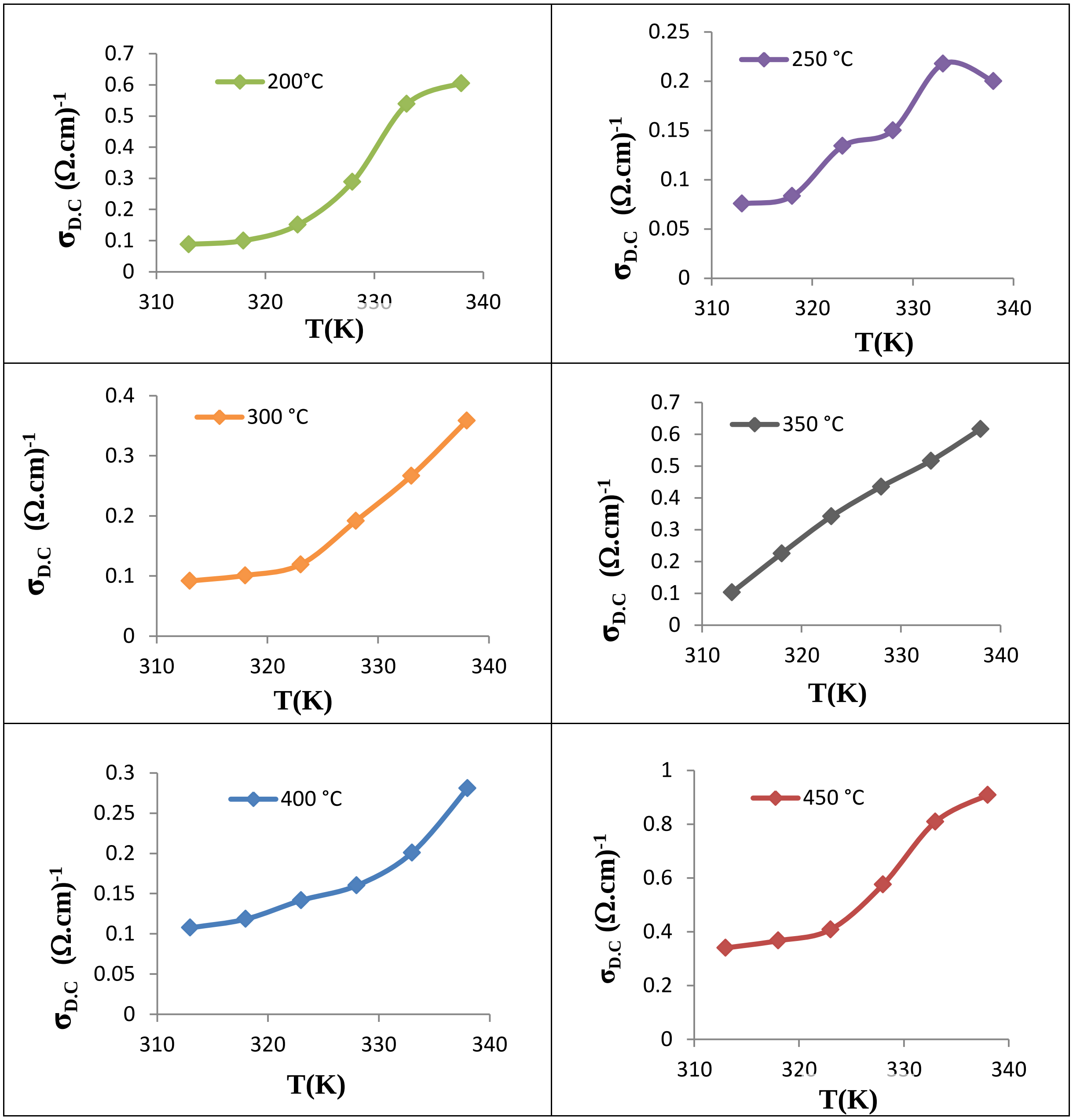

Fig.(1): Variation of $\sigma_{D.C}$ versus temperature for PbO films at different substrate temperatures (200, 250, 300, 350, 400, 450)˚C .

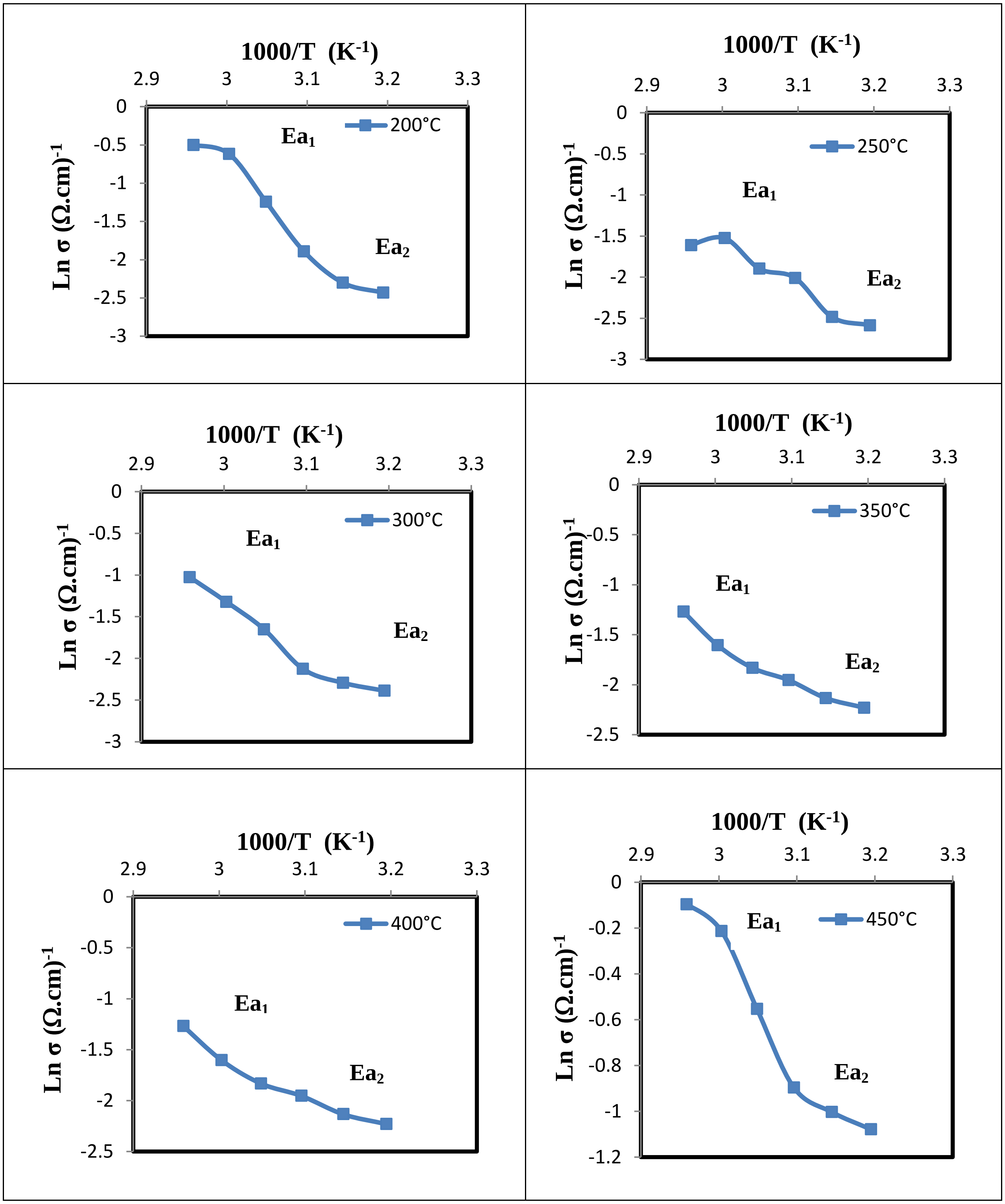

Fig.(2): Lpnσ versus 1000/T for (PbO) thin films at different substrate temperatures (200, 250, 300, 350, 400, 450)˚C

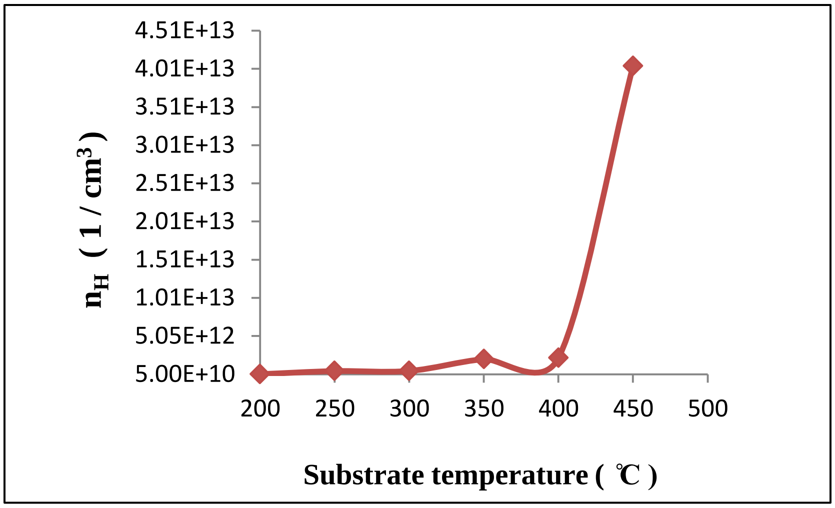

Fig.(3) : Variation of carrier concentration for PbO thin films with Substrate temperatures

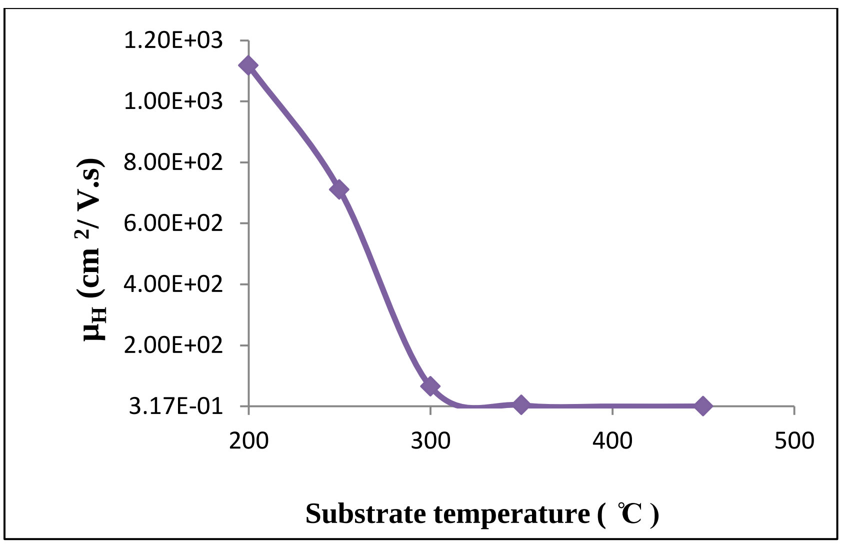

Fig.(4) : Variation of mobility for PbO thin films with Substrate temperature (200, 250, 300, 350, 400, 450)˚C

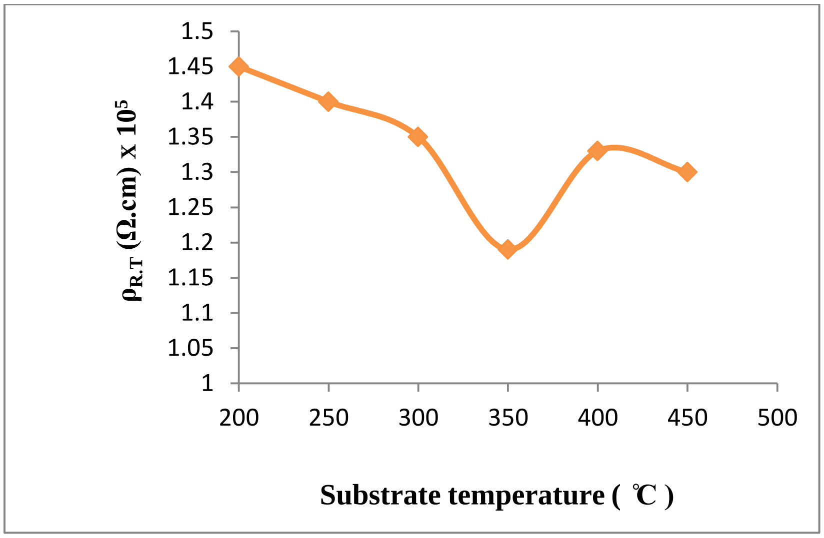

Fig.(5) : Variation of electrical resistivity for PbO thin films with Substrate temperatures.

Table (2): Hall parameters for (PbO) films at different substrate temperatures.

Substrate temperatures (℃)	R_H (cm^3 / C)	n_H (1 / cm^3)	$\sigma_{R.T} \times 10^{-6}$ $(\Omega.cm)^{-1}$	$\rho_{R.T} \times 10^5$ (Ω.cm)	μ_H (cm^2/ V.s)
200	$-2.16x10^8$	$-5.95x10^{10}$	6.85	1.45	$11.18x10^2$
250	$-1.63x10^7$	$-4.66\ x10^{11}$	8.32	1.40	$7.10\ x10^2$
300	$-1.44\ x10^7$	$-4.85\ x10^{11}$	7.22	1.35	$6.50\ x10^1$
350	$-2.37x10^6$	$-2.03x10^{12}$	4.56	1.19	4.08
400	$-5.15x10^5$	$-2.19x10^{12}$	7.47	1.33	$1.13\ x10^{-1}$
450	$-1.54x105^5$	$-4.04x10^{13}$	7.64	1.30	$3.17\ x10^{-1}$